

REMARKS

Claim 1 has been amended. Reexamination and reconsideration of the application as amended is requested.

The Examiner states that the cross-noting amendment to the specification after the title should be deleted as redundant. The reference has been deleted.

The Examiner rejected claim 1 under 35 U.S.C. § 102(b) for purported anticipation by or, in the alternative, under 35 U.S.C. § 103(a) for purported obviousness over U.S. Patent No. 5,322,888 to Kato et al. ("Kato"). The Examiner asserts that Kato discloses composite sols of Optolake 1130F-2 which is a $\text{SiO}_2/\text{TiO}_2/\text{Fe}_2\text{O}_3$ composite having a particle size of 20 nm in methanol, that the coating composition is surface modified with γ -glycidoxypolytriethoxysilane, and that the dielectric constant for methanol at 20°C is 31.2. The Examiner also asserts that the claimed molecular polarizability would have been inherent to the γ -glycidoxypolytriethoxysilane exemplified in the reference.

Claim 1 has been amended so that the ratio of silica to at least one inorganic oxide in the inorganic compound particulates is 3 to 500. The amendment is based on the maximum of the ranges provided on page 6, line 19 of the specification and the value provided in Example 2 at page 14, line 2. Therefore, no new matter is introduced by this amendment.

The present invention makes use of the dispersion of inorganic compound particulates in a dispersion medium. Particles of an inorganic oxide must generally have their surfaces rendered hydrophobic if they are to be dispersed in an organic solvent. Thus, generally, the surface of such particulates is modified.

A method of modifying the surface of such inorganic compound particulates comprises reacting, for example, a reactive monomer or a coupling agent with hydroxyl groups of the particulate surface. However, it is difficult to obtain an organic solvent sol of high

dispersibility. When these inorganic compound particulates are used as a filler in, for example, a coating material, a hard coating agent component of an insulating coat and a protective coat, adding a sol thereof to a matrix of coating film forming agent is likely to invite an aggregation of particulates in the matrix. In particular, problems have been encountered when an organic solvent sol has been used in the presence of a cation, anion or a surfactant; particulates may be aggregated and a gelation may occur. For example, an organic solvent sol in which inorganic compound particulates have been dispersed has been used in coating materials in order to improve hardness, water resistance and stain resistance of the resulting coating films. However, a problem has been encountered in that the coating material becomes viscous and whitens because the organic solvent sol has limited compatibility with the coating film-formed resin and the resin emulsion obtained.

In the present invention, SiO₂-rich composite particles are modified by an organosilane selected from vinylsilane compounds, acrylsilane compounds, epoxysilane compounds, aminosilane compounds, γ -mercaptopropyltrimethoxysilane and γ -chloropropyltrimethoxysilane, exhibiting a molecular polarizability of 2×10^{-40} to 850×10^{-40} C²m²J⁻¹, wherein the SiO₂ composite particles are composite oxide particles composed of silica and at least one inorganic oxide other than silica.

In the inorganic compound sol of the present invention, the surface of the SiO₂ composite particles is modified by an organosilane selected from vinylsilane compounds, acrylsilane compounds, and an epoxysilane organic compound having a specified molecular polarizability. Therefore, the SiO₂ composite particles have a desirable affinity with the dispersion media, so that the dispersion stability of the SiO₂ composite particles in the dispersion media is excellent, and the aggregation of particles and gelation do not occur. Further, even if an organic

or inorganic acid or a salt thereof were present in the inorganic compound sol, the SiO₂ composite particles would not aggregate with each other and the gelation would not occur.

When the inorganic compound sol of the present invention is blended as a filler in, for example, a coating material or a hard coating agent even if an acid, base, salt, surfactant, et al. is present in the inorganic compound sol, the resulting coating film is substantially free from decrease of transparency and cracking attributed to the aggregation of particulates and gelation. Therefore, the inorganic compound sol of the present invention is useful as a filler to be blended in not only various coating materials and hard coating agents but also various resins.

Kato contains a description of a coating composition for optical plastic moldings comprising a hydrolyzate of a silane compound, titanium oxide-based composite fine particles, an unsaturated or saturated polycarboxylic acid or anhydride thereof, and a heat-curing catalyst (see claim 1).

The differences between Kato and the present invention are shown in the following

Table 1:

	Present Invention	U.S. Patent No. 5,322,888 (Kato)
Inorganic compound particulates	Silica-based composite particles	Titanium oxide-based composite particles
Ratio of silica to other inorganic	3 to 500	0.03 to 0.7 SiO ₂ /(TiO ₂ +Fe ₂ O ₃) = 10/90 (0.11) in Example 7
Surface modifier of particles	vinylsilane compounds, acrylsilane compounds, epoxysilane compounds, aminosilane compounds, γ -mercaptopropyltrimethoxysilane, exhibiting a molecular polarizability of 2×10^{-40} to $850 \times 10^{-40} \text{ C}^2 \text{ m}^2 \text{ J}^{-1}$.	tetramethoxysilane, methyltrimethoxysilane, trimethylchlorosilane, vinyltriethoxysilane, γ -glycidoxypentyltriethoxysilane, γ -glycidoxypentylmethyldiethoxysilane
Hydrolyzate of a silane compound (matrix)	Present in small amounts only	Present
Unsaturated or saturated	Not present	Present

polycarboxylic acid or anhydride		
Heat-curing catalyst	Not present	Present

(1) The particles used in the present invention differ from the particles used by Kato. In the present invention, the SiO₂ composite particles are composite oxide particles composed of silica and at least one other inorganic oxide other than silica, wherein the silica content is high. In contrast, the fine particles used by Kato are a titanium oxide-based composite in which the titanium oxide content is high and the silica content is far less than in the present invention. Amended claim 1 of the present application points out this distinction.

(2) In the present invention, the SiO₂ composite particles are modified by an organosilane selected from vinylsilane compounds, acrylsilane compounds, epoxysilane compounds, aminosilane compounds, γ -mercaptopropyltrimethoxysilane and γ -chloropropyltrimethoxysilane, exhibiting a molecular polarizability of 2×10^{-40} to $850 \times 10^{-40} \text{ C}^2 \text{ m}^2 \text{ J}^{-1}$. Kato teaches surface modification by, for example, dipping fine particles in an alcohol solution of the silane coupling agent. An unselective common silane coupling agent is used as a surface modifier. In the present invention, organosilanes characterized by both chemical composition and a molecular polarizability of 2×10^{-40} to $850 \times 10^{-40} \text{ C}^2 \text{ m}^2 \text{ J}^{-1}$ are used as surface modifiers of the SiO₂ rich composite particle. Kato neither suggests nor discloses the use of a specific organosilane exhibiting a molecular polarizability of 2×10^{-40} to $850 \times 10^{-40} \text{ C}^2 \text{ m}^2 \text{ J}^{-1}$ as a surface modifier of a SiO₂ rich composite particle.

The embodiment of the present invention is further distinguished from Kato by being a sol in which the SiO₂ rich composite particles are stably dispersed. The sol is used as a

filler for coating material, hard coating agent and cement. In contrast, Kato makes no reference to a sol in which the composite particles are stably dispersed. The embodiment of the Kato patent is the coating composition in which the titanium oxide-based composite fine particles modified by silane coupling agent are contained. The titanium oxide-based composite fine particles modified by a silane coupling agent of Kato are used together with a hydrolyzate of a silane compound, an unsaturated or saturated polycarboxylic acid or an anhydride thereof and a heat-curing catalyst of the silane compound. Furthermore, Kato is concerned with a coating composition for optical plastic moldings. The Kato composition forms a coating film having resistance to weathering, impact, heat, hot water, chemicals, and wear, and good clarity, flexibility, dyeability, and adhesion to deposited metal film. Therefore, the application of the present invention is different from that of Kato. The sol of the present invention does not contain a hydrolyzate of a silane compound, an unsaturated or saturated polycarboxylic acid or an anhydride thereof, or a heat-curing catalyst of the silane compound.

(3) The modification of the surface of the SiO_2 rich composite particles by an organosilane of a type claimed in claim 1, with a molecular polarizability claimed in claim 1, makes the SiO_2 rich composite particles have a desirable affinity with the dispersion media, so that a highly stable dispersion of the particles in the dispersion media is produced, and aggregation of particulates and gelation do not result. Even if an organic or inorganic acid or a salt thereof is present in the inorganic compound sol, the SiO_2 rich composite particles would not aggregate with each other and gelation would not occur. This effect is clear from a comparison of Example 1 and Comparative Example 2 in Table 2.

Table 2

	Sol Composition	Modifying organic compound		Dispersion medium	Stability	
					Sulfuric Acid	Hydrochloric acid
Ex. 1	SiO ₂ · Al ₂ O ₃	γ-glycidoxy-propyltrimethoxysilane	10.4 x 10 ⁻⁴⁰	Ethylene glycol	Stable at least 6 months	Stable at least 6 months
Comp. Ex. 2	SiO ₂ · Al ₂ O ₃	Monomethyltrimethoxysilane	1.9 x 10 ⁻⁴⁰	Ethylene glycol	Gelled 30 days	Gelled 30 days

As is shown in Table 2, when the modifying organic compound has a molecular polarizability higher than $2 \times 10^{-40} \text{ C}^2 \text{ m}^2 \text{ J}^{-1}$, the resulting organic compound sol has excellent dispersibility in an organic solvent even if inorganic acids are present in the sol. (See Example 1.) In contrast, when the modifying organic compound has a molecular polarizability lower than $2 \times 10^{-40} \text{ C}^2 \text{ m}^2 \text{ J}^{-1}$, the resulting organic compound sol has poor dispersibility in an organic solvent. (See Comparative Example 2.) This effect is not suggested by Kato. As mentioned previously, the coating composition of Kato apparently differs from that of the present invention. Therefore the effect of the particulates of the present invention is neither disclosed nor suggested by Kato.

The Examiner rejected claim 1 under 35 U.S.C. § 102(e) as anticipated by or, in the alternative, under 35 U.S.C. § 103(a) as obvious over U.S. Patent No. 5,858,077 to Kayanoki ("Kayanoki"). The Examiner asserts that Kayanoki discloses compositions comprising a composite metal oxide dispersed in methanol and/or isopropanol with the further addition of a polymerizing monomer. The Examiner also asserts that Kayanoki teaches particle sol compositions (Optolake 1130 F-2 (A-8)) wherein the particles have a particle size of 10 nm and broadly may be 1 to 100 nm., and teaches treating the methanol dispersed composite particle sol

with γ -glycidoxypyrtrimethoxysilane followed by the addition of isopropanol and a polymerization monomer. The Examiner further asserts that the claimed dielectric constant property of the dispersion medium would have been expected to have been inherent to the dispersion medium disclosed in Kayanoki.

Kayanoki describes a coating composition comprising (1) fine particles of at least one oxide selected from the group consisting of silica, iron oxide, titanium oxide, cerium oxide, zirconium oxide, antimony oxide, zinc oxide and tin oxide, and composite oxides thereof; (2) an epoxy group-containing silicon compound or a partial hydrolyzate thereof; and (3) an organic compound having one OH group or SH group in the molecule and containing in the main chain thereof at least one bonding unit selected from the group consisting of $-\text{O}-$, $-\text{CO}-\text{O}-$, $-\text{S}-$, $-\text{CO}-\text{S}-$ and $-\text{CS}-\text{S}-$ along with at least one unsaturated group, the organic compound being soluble in water or a lower alcohol having up to 4 carbon atoms.

Kayanoki never discloses that the silica rich composition particle has a ratio of silica to at least one organic oxide other than silica of 3 to 500 as is claimed in the amended claim 1. Kayanoki teaches a ratio of silica to other inorganic oxides of 0.001 to 1. The silica content of the Kayanoki composite particle is far less than that use in the present invention. In addition, it is stated at col. 3, lines 10-11 that particles with the composition of those in the present invention are unsuitable for use in the Kayanoki formulation. According to Kayanoki, a ratio of SiO_2 and $(\text{CeO}_2 + \text{TiO}_2)$ exceeding 0.5 produces a dispersion with an undesirably low stability. Kayanoki cannot teach the modification of particles by the organic compounds of the present invention if the use of these particles is specifically contraindicated.

The amount of the epoxy group-containing silicon compound or partial hydrolyzate thereof in the Kayanoki coating composition is much greater than the amount of organosilane

compound used in the present invention. In the present invention, the amount of organosilane compound is very small. For example, 8.4 g of the organosilane compound are used in combination with 90 g of SiO₂ composite particles (9.3% by weight) in Example 1 of the present invention. In contrast, the epoxy group-containing silicon compound or a partial hydrolyzate thereof is used in an amount of from 5 to 90 wt % based on the total solids in the coating composition, and the fine particles are used in an amount of 10% to 70%, based on the total solids in the coating composition of Kayanoki. The amounts of the epoxy group-containing silicon compound or a partial hydrolyzate used by Kayanoki indicate that the compound is used as a coating forming component (matrix) and is not used for modifying the oxide particles. Kayanoki does teach, as was noted previously, that dispersion stability depends on the composition of the oxide particles. Use of the compound as a coating forming component for silica-poor particles by Kayanoki does not suggest its use for modifying the surface of silica-rich particles in the present invention.

Kayanoki describes subjecting fine particles to surface modification with organosilicon compounds. Many and various organosilicon compounds are listed as examples (see column 3, line 19 through column 4, line 18). However the selective use of specific organic compounds selected from vinylsilane compounds, acrylsilane compounds, epoxysilane compounds, aminosilane compounds, γ -mercaptopropyltrimethoxysilane and γ -chloropropyltrimethoxysilane, exhibiting a molecular polarizability of 2×10^{-40} to 850×10^{-40} C² m² J⁻¹ as a silane coupling agent for particles with the SiO₂ content of the particles of the present invention is not described by Kayanoki. As mentioned previously, the modification of the surface of the SiO₂ composite particles by an organosilane, selected from vinylsilane compounds, acrylsilane compounds, and epoxysilane compounds, having a specified molecular polarizability

produces SiO₂ composite particles having the desired affinity with the dispersion media, so that the desired dispersion stability of the particles in the media is produced, and aggregation of particulates and gelation do not occur. In addition, even if an organic or inorganic acid or a salt thereof is present in the organic compound sol, the SiO₂ composite particles would not aggregate with each other and gelation would not occur. This effect cannot be suggested by Kayanoki, which does not disclose the selective use of specific organic compounds exhibiting a molecular polarizability of 2×10^{-40} to $850 \times 10^{-40} \text{ C}^2 \text{ m}^2 \text{ J}^{-1}$ for particles in which silica predominates.

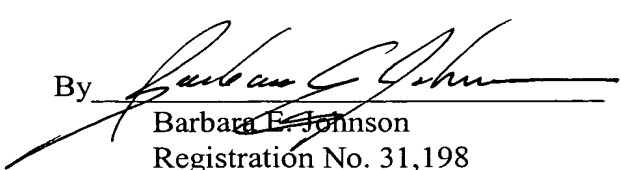
For these reasons, the particles and organic compounds of Kayanoki differ from those of the present invention and, therefore, the sol of the present invention is neither disclosed nor suggested by Kayanoki. Accordingly, it is believed that claim 1 is neither anticipated by nor obvious over Kayanoki.

In view of the above, it is submitted that the claim is in condition for allowance. Reconsideration of the rejections is requested. Allowance of claim 1 at an early date is solicited.

Respectfully submitted,

WEBB ZIESENHEIM LOGSDON
ORKIN & HANSON, P.C.

By



Barbara E. Johnson
Registration No. 31,198
Attorney for Applicant
700 Koppers Building
436 Seventh Avenue
Pittsburgh, PA 15219-1818
Telephone No. 412-471-8815
Facsimile No. 412-471-4094



MARKED UP AMENDED CLAIM 1

1. (Thrice Amended) An inorganic compound sol comprising a dispersion medium having a dielectric constant of from 10 to 85 and, dispersed therein, inorganic compound particulates having average particle size from about 11 to about 30 nm whose surface has been modified by an organic compound which is selected from the class consisting of vinylsilane compounds, acrylsilane compounds, epoxysilane compounds, aminosilane compounds, γ -mercaptopropyltrimethoxysilane and γ -chloropropyltrimethoxysilane, exhibiting a molecular polarizability of from 2×10^{-40} to $850 \times 10^{-40} \text{ C}^2\text{m}^2\text{J}^{-1}$, wherein the inorganic compound particulates are composite oxide particulates composed of silica and at least one inorganic oxide other than silica, with the ratio of silica to at least one inorganic oxide other than silica being 3 to 500.